# *Studies of thermal transport in Mo/Si multilayer structures*

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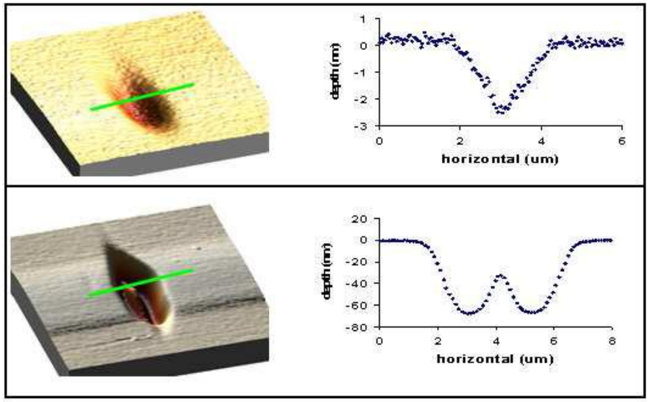
# Heat Loads on multilayer optics

- *High heat loads - radiation generated damage*




Sobierajski et al, EUV Source Workshop, Dublin (2014)

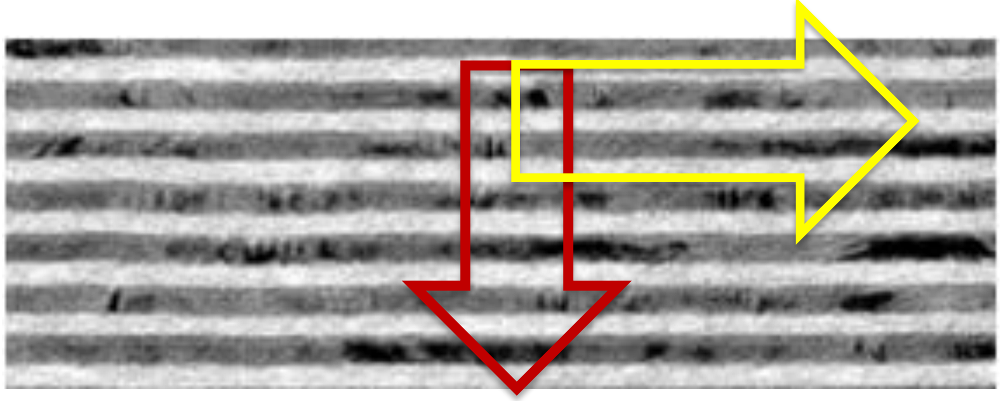
- *Moderate heat loads - heat induced imaging distortions*

*!To be investigated*

# Heat transport in Mo/Si multilayers

Anisotropic heat transport

Lateral transport driven by electrons

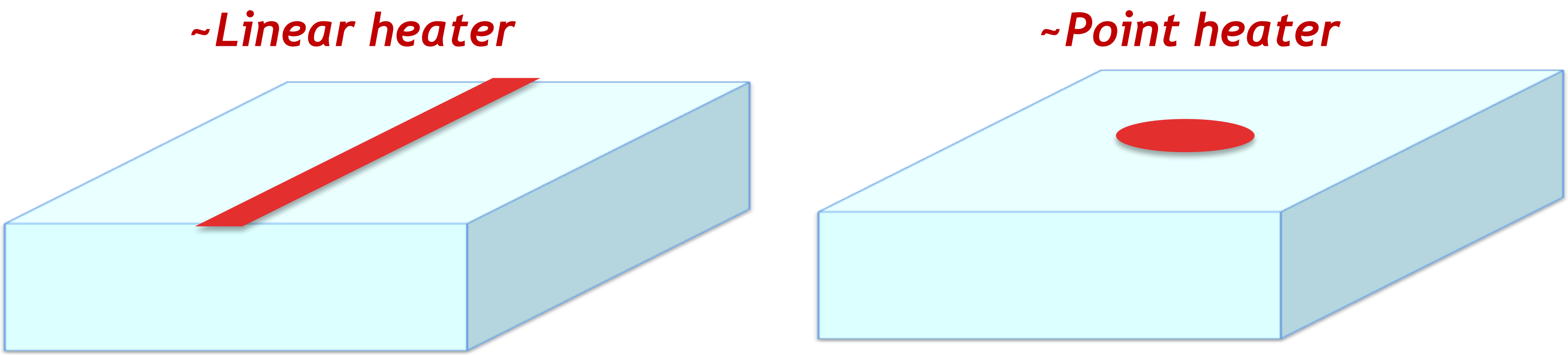


In-depth transport driven by lattice vibrations (phonons)

But how to characterize it?

# Techniques utilizing thermal waves

- Well-determined modulated heat source is applied on sample (thin film) surface

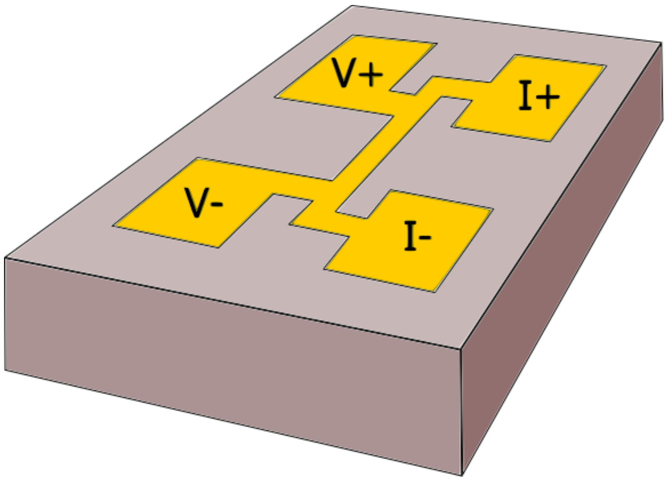



- Modulated heat source induces thermal wave propagation in sample volume

- Oscillation of surface temperature is measured to characterize thermal properties of the sample

# 3ω techinque

*Geometry for thermal conductivity (k) measurements using 3ω technique*




*Idea of 3ω technique*

*Current:*

**I ~ cos(ωt)**

*Power dissipation:*

**P ~ $I^2$ ~ cos(2ωt)**

*Temperature* **ΔT(ω)** *induced resistance oscillations:*

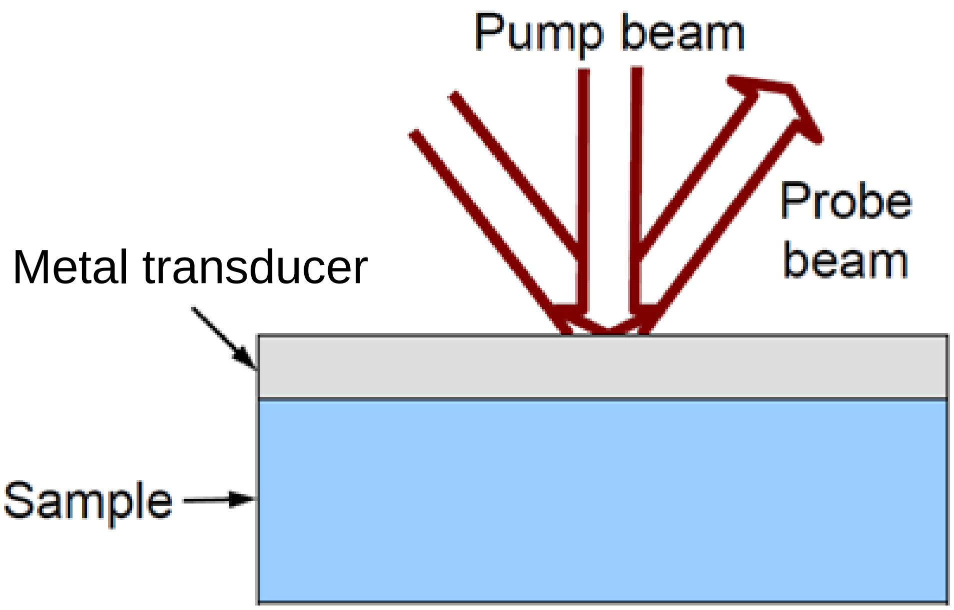
**ΔR ~ cos(2ωt)**

*Small voltage oscillations at 3ω:*

**ΔV ~ I·ΔR~cos(3ωt)** **- main observable**


D. G. Cahill *Rev. Sci. Instrum.* **61**, 802 (1990)

# Transient thermoreflectance




*Probe beam heats sample surface*:

$\Delta T$

*Optical constants and reflectivity depend on temperature*

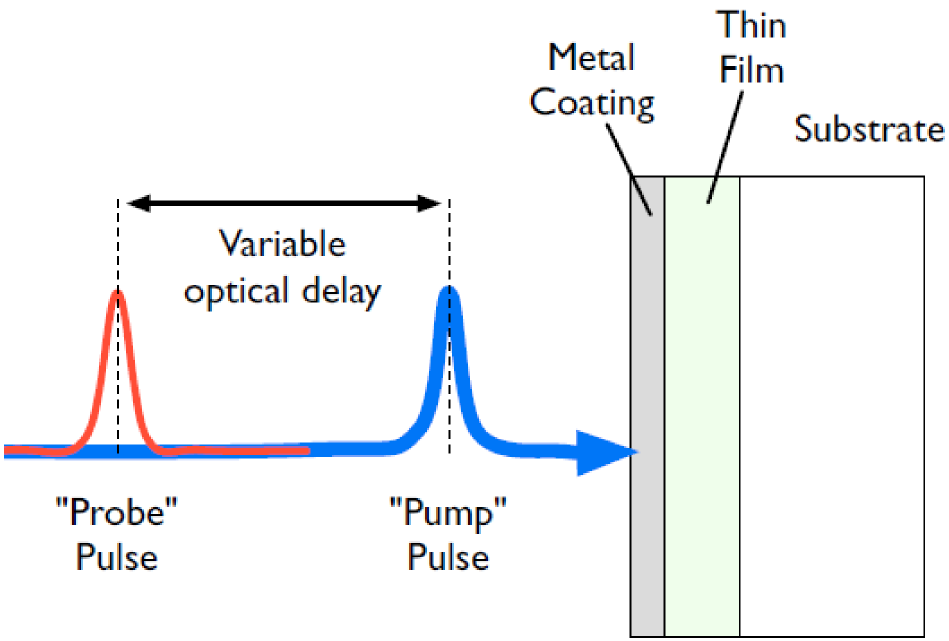
$\Delta T \rightarrow \Delta R$

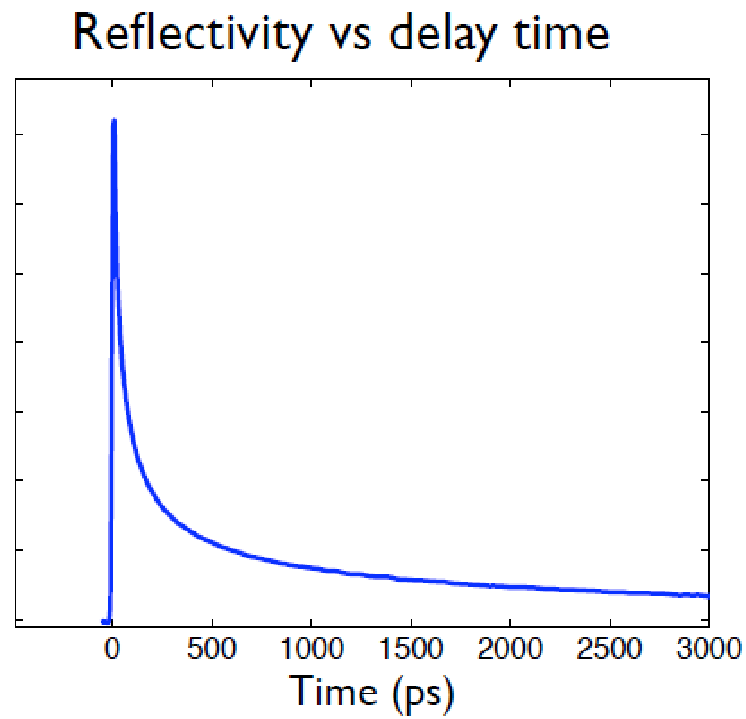
*Probe beam measures reflectivity changes*

$\Delta R_{meas} \rightarrow \Delta T_{meas}$

D. Cahill, Review of scientific instruments 75 (12), 5119-5122 (2004)
A.J.Schmidt et al, Review of Scientific Instruments 79 (11), 114902 (2008)
A.J.Schmidt et al, Review of Scientific Instruments 80 (9), 094901 (2009)

# Time Domain ThermoReflectance (TDTR)

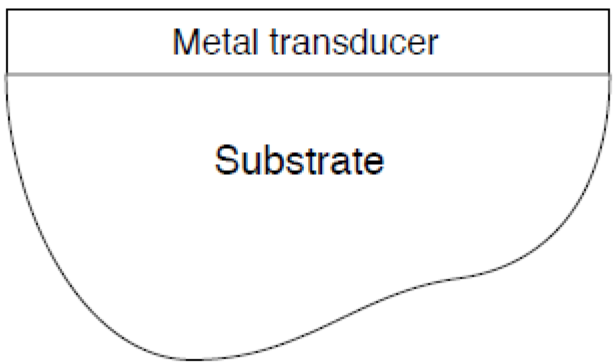


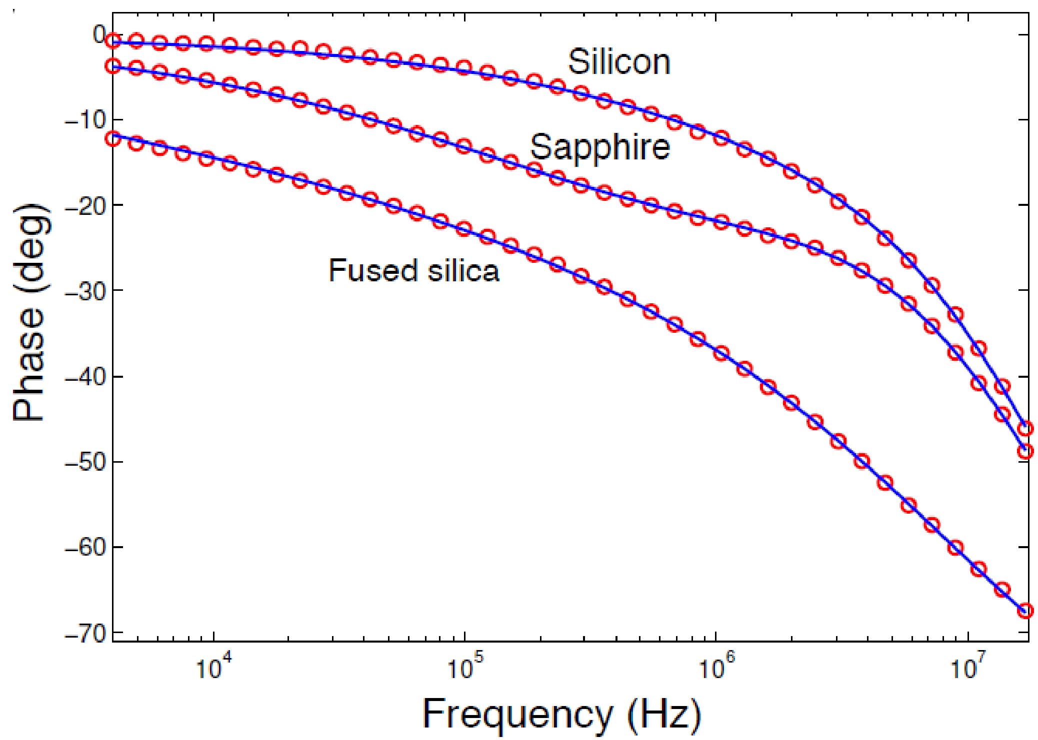



# Frequency Domain ThermoReflectance (FDTR)

$$qe^{i\omega t} \longrightarrow \boxed{H(r,z,\omega)} \longrightarrow T(r,z,\omega)e^{i[\omega t+\phi(r,z,\omega)]}$$

Periodic heat source — Frequency response — Surface temperature

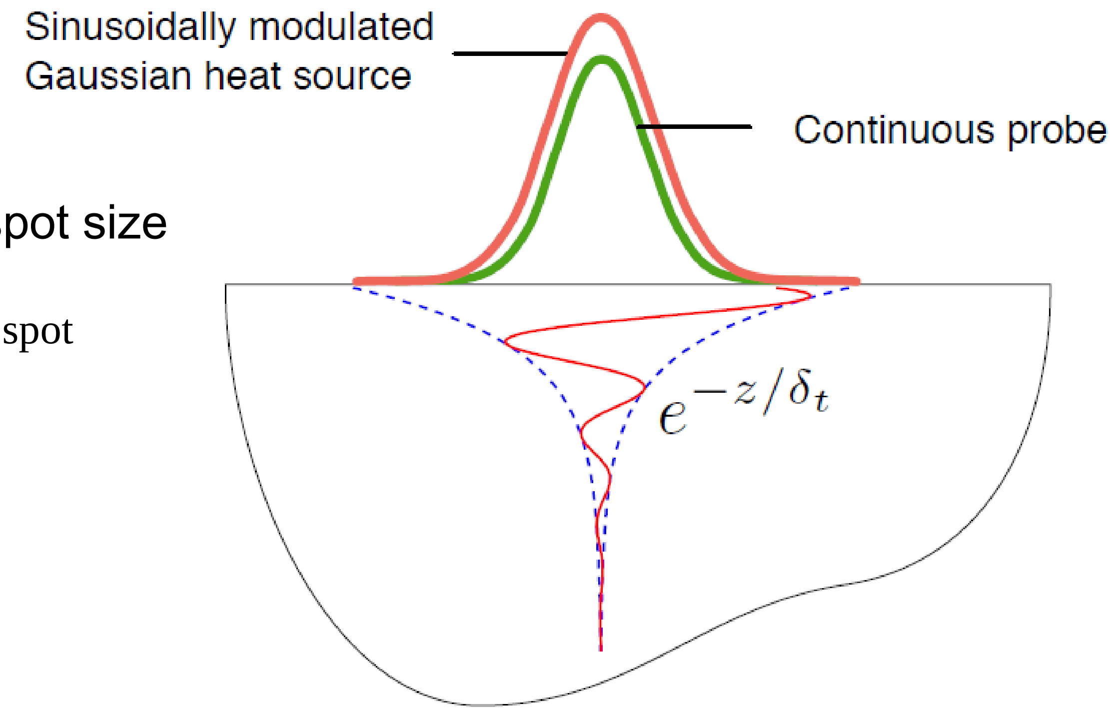


# Thermal wave penetration depth

Laser spot size

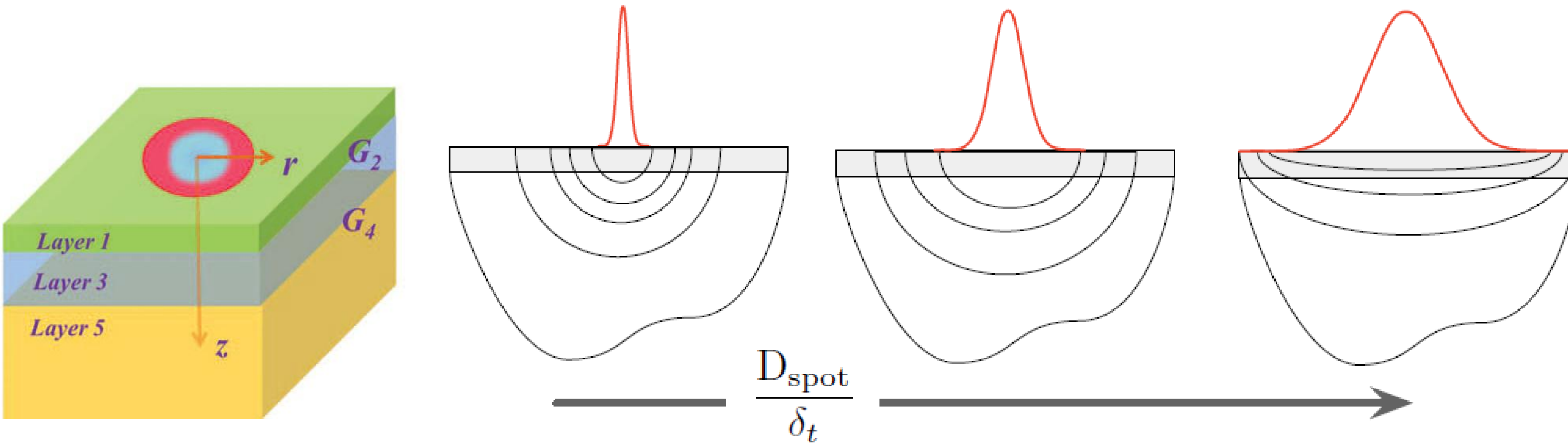
$D_{spot}$




$$\delta_t = \sqrt{\frac{2\kappa}{C\omega}}$$

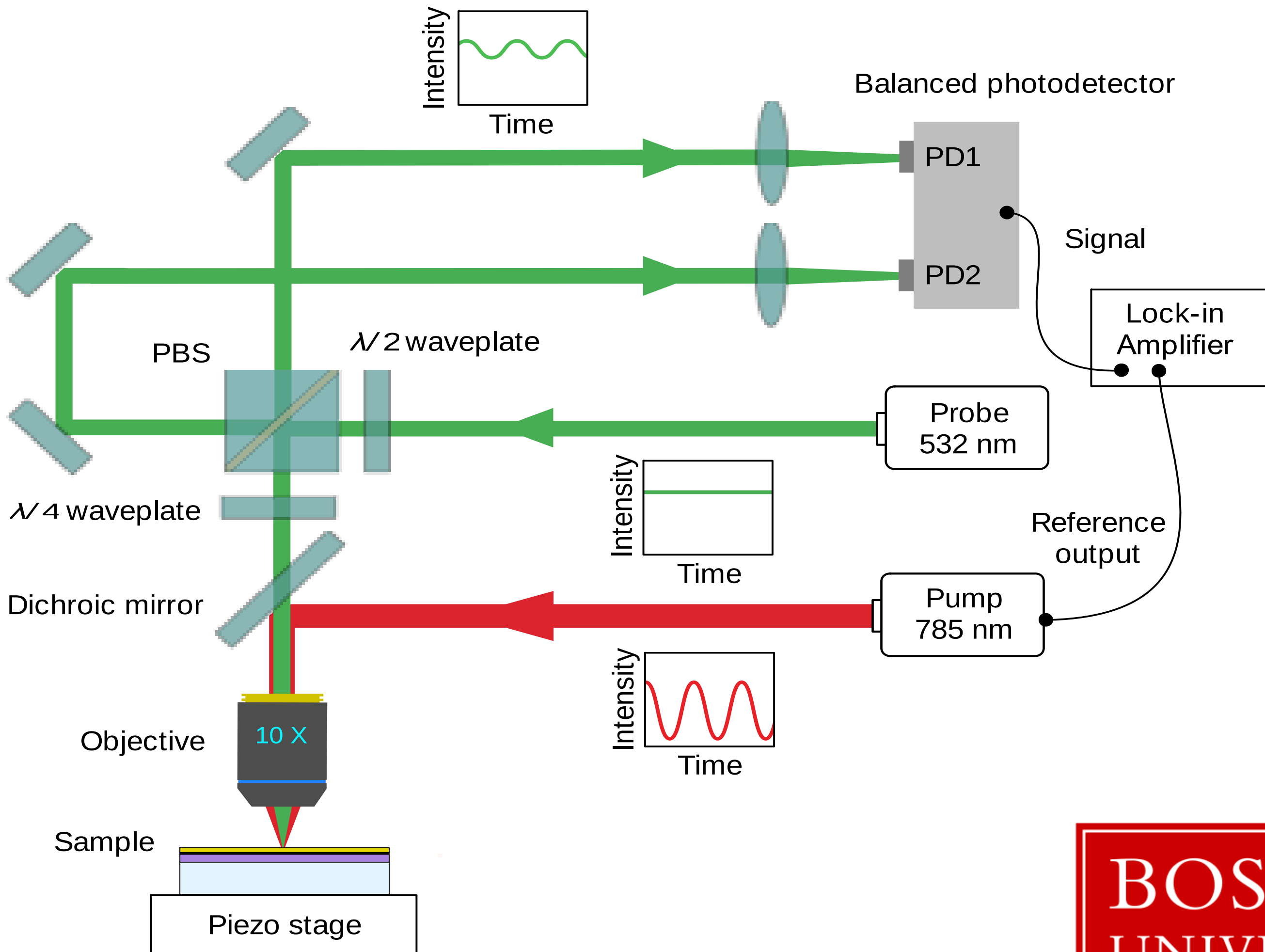
$D_{spot}/\delta_t$ ratio determines ‘geometry’ of thermal wave propagation

# Switching between 1D and 3D heat conduction




| | | | |
|---|---|---|---|
| Thermal wave solution | Quasi-steady three-dimensional spherical wave | | One-dimensional plane wave |
| $\omega$ | $\omega \ll \frac{32\kappa_{r3}}{R_{pp}C_3}$ | $\omega \sim \frac{32\kappa_{r3}}{R_{pp}C_3}$ | $\omega \gg \frac{32\kappa_{r3}}{R_{pp}C_3}$ |
| Thermal property | $\sqrt{\kappa_{z3}\kappa_{r3}}$ | $\sqrt{\kappa_{z3}\kappa_{r3}}, \sqrt{\kappa_{z3}C_3}$ | $\sqrt{\kappa_{z3}C_3}$ |

# FDTR setup

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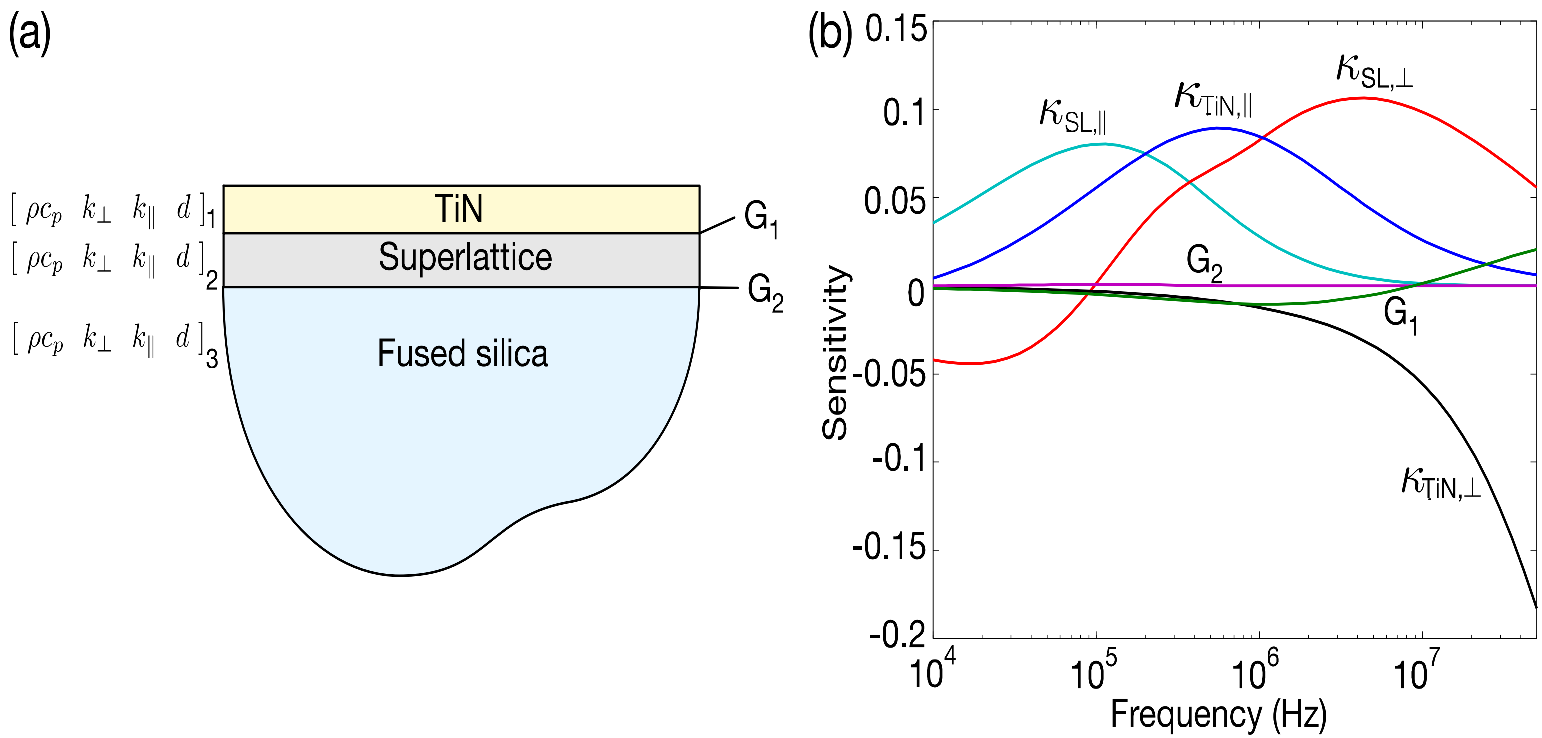
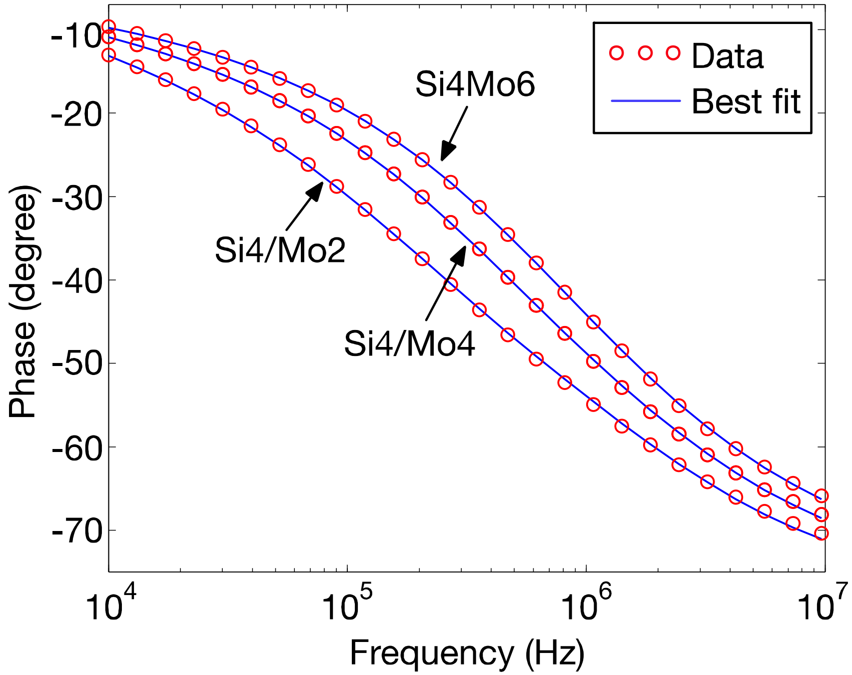
# Parameters sensitivity



# Transient thermoreflectance

## Measured FDTR data




## Multilayer properties obtained from data fit

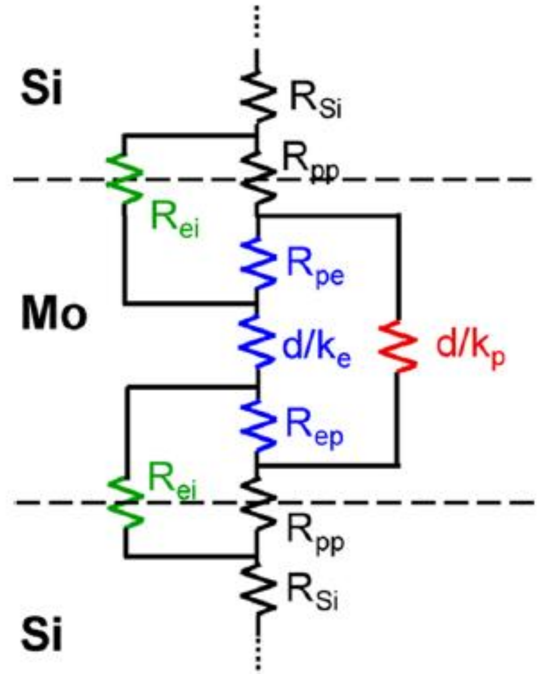
| Sample | $k_{cross\text{-}plane}$ ($W\ m^{-1}K^{-1}$) | $k_{in\text{-}plane}$ ($W\ m^{-1}K^{-1}$) |
|---|---|---|
| Si4/Mo2 | 0.75 ± 0.03 | 2.47 ± 0.14 |
| Si4/Mo4 | 1 ± 0.03 | 8.48 ± 0.18 |
| Si4/Mo6 | 1.3 ± 0.03 | 15.7 ± 0.24 |

# Cross-plane

$$R_{Mo/Si} = \frac{d_{Mo} + d_{Si}}{k_{\perp}} = \frac{d_{Mo}}{k_{Mo}} + \frac{d_{Si}}{k_{Si}} + \frac{1}{G_{Si-Mo}} + \frac{1}{G_{Mo-Si}}$$

*k* - thermal conductivity

*G* - thermal boundary conductance

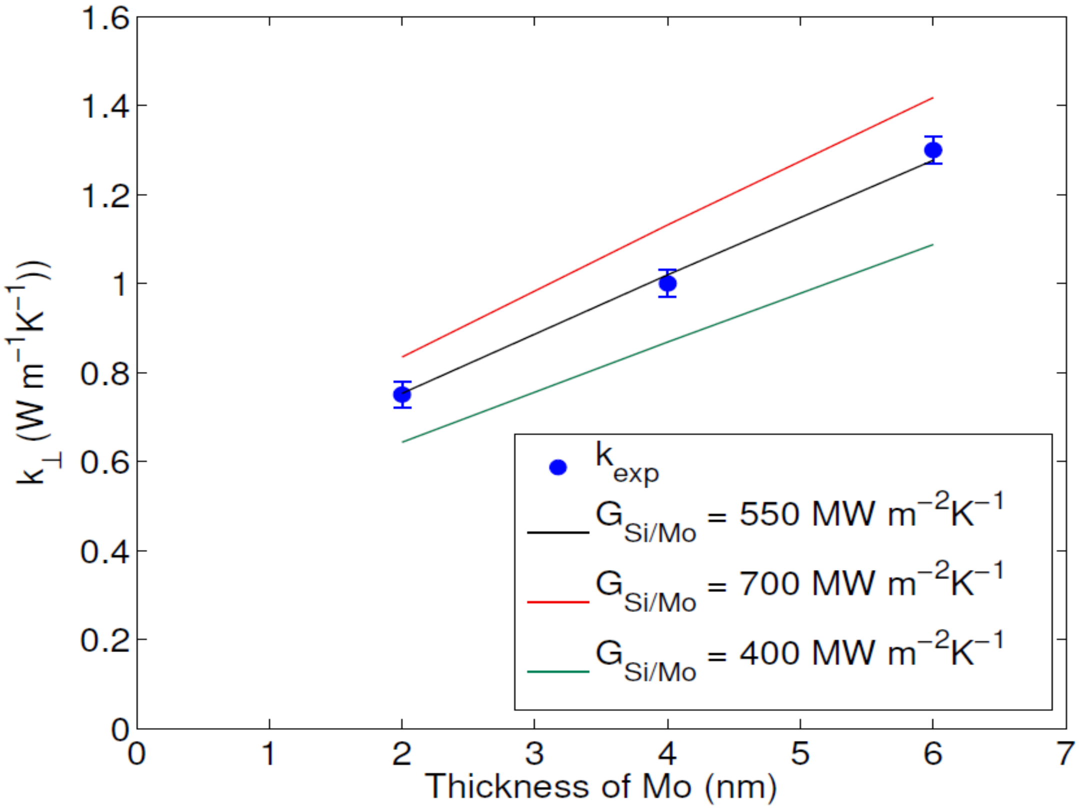


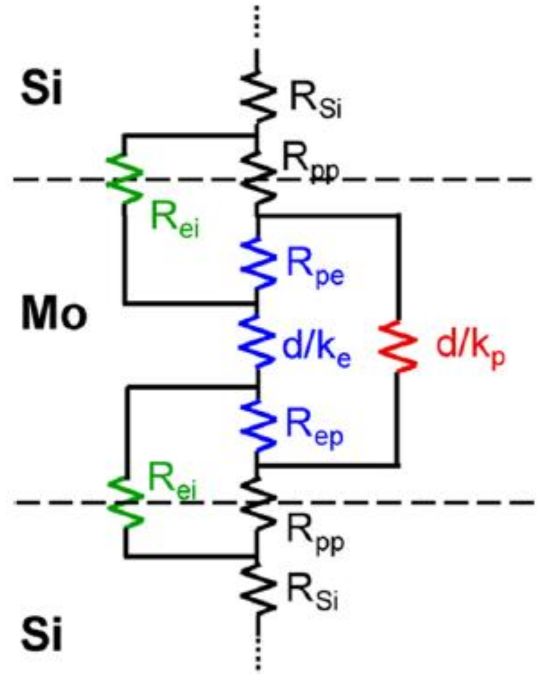

| Material | *k* (W/m-K) |
|---|---|
| c-Si | 149 |
| a-Si | **1** |
| Mo | 138 |

Low cross-plane k values for Mo/Si due to a-Si and interface scattering

# Cross-plane

$$R_{Mo/Si} = \frac{d_{Mo} + d_{Si}}{k_{\perp}} = \frac{d_{Mo}}{k_{Mo}} + \frac{d_{Si}}{k_{Si}} + \frac{1}{G_{Si-Mo}} + \frac{1}{G_{Mo-Si}}$$

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# Analysis of in-plane thermal conductivity

| Sample | $k_{in\text{-}plane}$ ($W\ m^{-1}K^{-1}$) |
|---|---|
| Si4/Mo2 | 2.47 ± 0.14 |
| Si4/Mo4 | 8.48 ± 0.18 |
| Si4/Mo6 | 15.7 ± 0.24 |

Values of in-plane thermal conductivity are relatively small

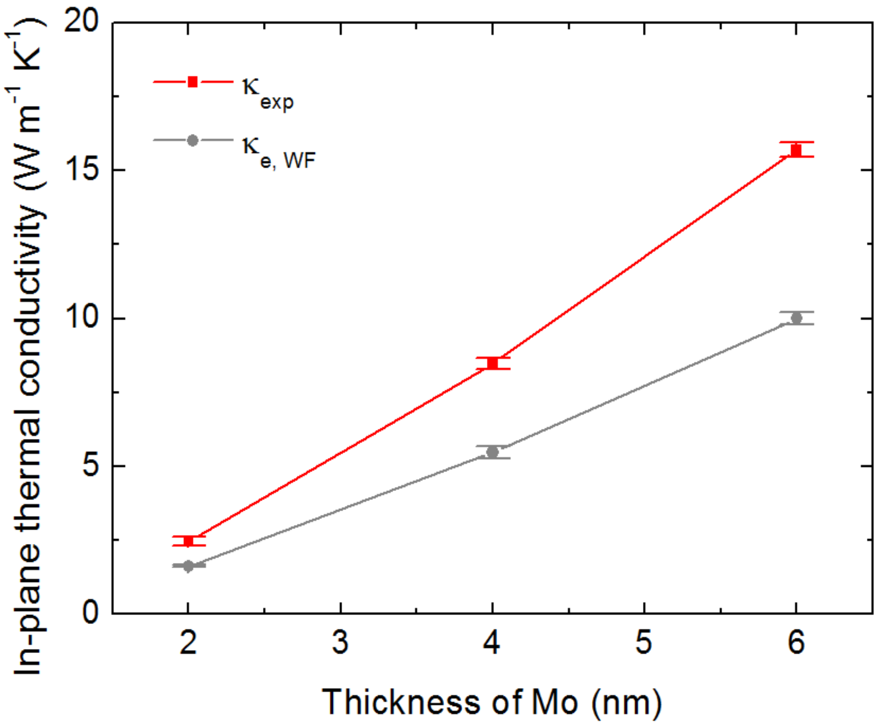
E.g. a-Si has $k \approx 1.0\ Wm^{-1}K^{-1}$.

- Lattice (phonon) contribution is significant
- Both contributions have to be analyzed

$$k = k_e + k_p$$

# In-plane thermal conductivity

Electron contribution estimated using electrical conductivity and the Wiedemann-Franz law




$$\frac{k}{\sigma} = LT$$

- Phonons contribute about 35% to in-plane heat conduction

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# Summary

- FDTR technique was adopted to characterize anisotropy of heat conduction in Mo/Si multilayer optics

- Cross-plane thermal transport governed by lattice vibrations. Small k values due to amorphous silicon and interface scattering

- In-plane thermal transport dominated by electrons, but lattice contribution is also considerable unlike in bulk metals

- Anisotropy ratio ($k_{in\text{-}plane}/k_{cross\text{-}plane}$) ranging 3.3 to 12.1 demonstrated for Mo/Si with varied Mo thickness from 2 to 4 nm, respectively

# Thank you for your attention

# Wiedemann-Franz law

WF law is the ratio of the electronic contribution of the thermal conductivity ($\kappa$) to the electrical conductivity ($\sigma$) of a metal, and is proportional to the temperature ($T$):

$$\frac{k}{\sigma} = LT$$

Theoretical value for Lorenz number (! No account for electron-phonon coupling):

$$L_0 = \frac{\pi^2}{3}\left(\frac{k_B}{e}\right)^2 = 2.44\times10^{-8}\left[\mathrm{W\Omega K^{-2}}\right]$$

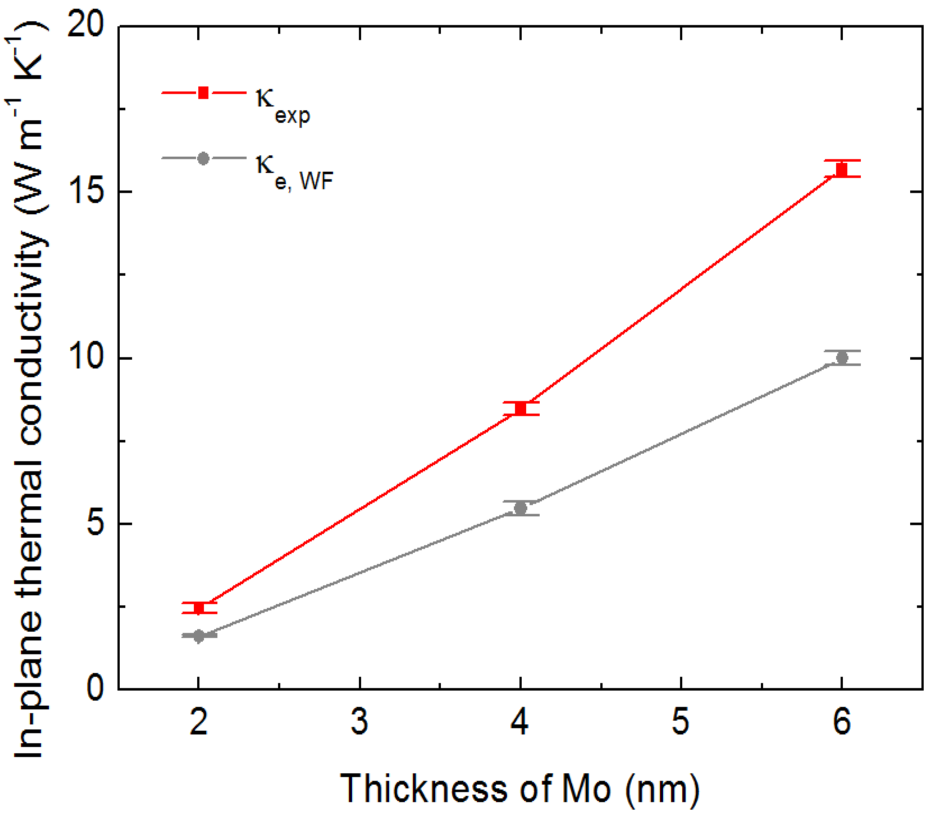
Corrected Lorenz number (Stojanovic et al *PRB* **82** 075418 (2010)) :

$$L_c = L_0\left[1+\frac{3}{\pi^2}\left(\frac{2}{Z}\right)^{2/3}\left(\frac{\theta_D}{T}\right)^2\right]^{-1}$$

# Four-probe measurement of electrical conductivity

| Sample | Number of periods | Si layer (nm) | Mo layer (nm) | $\sigma$ $10^6\ \Omega^{-1}m^{-1}$ |
|---|---|---|---|---|
| Si4/Mo2 | 160 | 4 | 2 | $0.312 \pm 0.005$ |
| Si4/Mo4 | 120 | 4 | 4 | $1.043 \pm 0.044$ |
| Si4/Mo6 | 96 | 4 | 6 | $1.898 \pm 0.040$ |

# In-plane thermal conductivity




- Phonons contribute about 35% to in-plane heat conduction

# How to estimate phonon contribution

Assumption of zero silicide layers, i.e. perfect Mo/Si structure:

$$k_p = \Gamma k_{p,Mo} + (1-\Gamma) k_{p,Si}$$

$$\Gamma = \frac{d_{Mo}}{d_{Mo} + d_{Si}}$$

We used measured value for a-Si:

$k = 1.01\ \mathrm{Wm^{-1}K^{-1}}$ (FDTR for 500 nm sputtered Si)

For Mo phonon contribution cannot be measured, needs to be estimated

# How to estimate phonon contribution

General formulae for any type of heat carriers:

$$k = \frac{1}{3} C_V v \lambda$$

$C_V$ - heat capacity, $v$ - velocity of heat carriers, $\lambda$ - mean free path

Phonons:

Heat capacity can be estimated using the Debye theory

$$C_V = 9Nk_B \left( \frac{T}{\theta_D} \right)^3 \int_0^{\theta_D/T} \frac{x^4 e^x}{\left(e^x - 1\right)^2} dx$$

Velocity of phonons equals speed of sound

$$v_S = \left(v_l^{-3} + 2v_t^{-3}\right)^{-1/3}$$

# Phonon mean free path

Two estimates are possible here

(Terry M. Tritt *Thermal conductivity: theory, properties and applications* 2004)

1. Minimum thermal conductivity limit

For highly disordered (amorphous) media phonon mean free pass is set to interatomic distance

$$\lambda = a$$

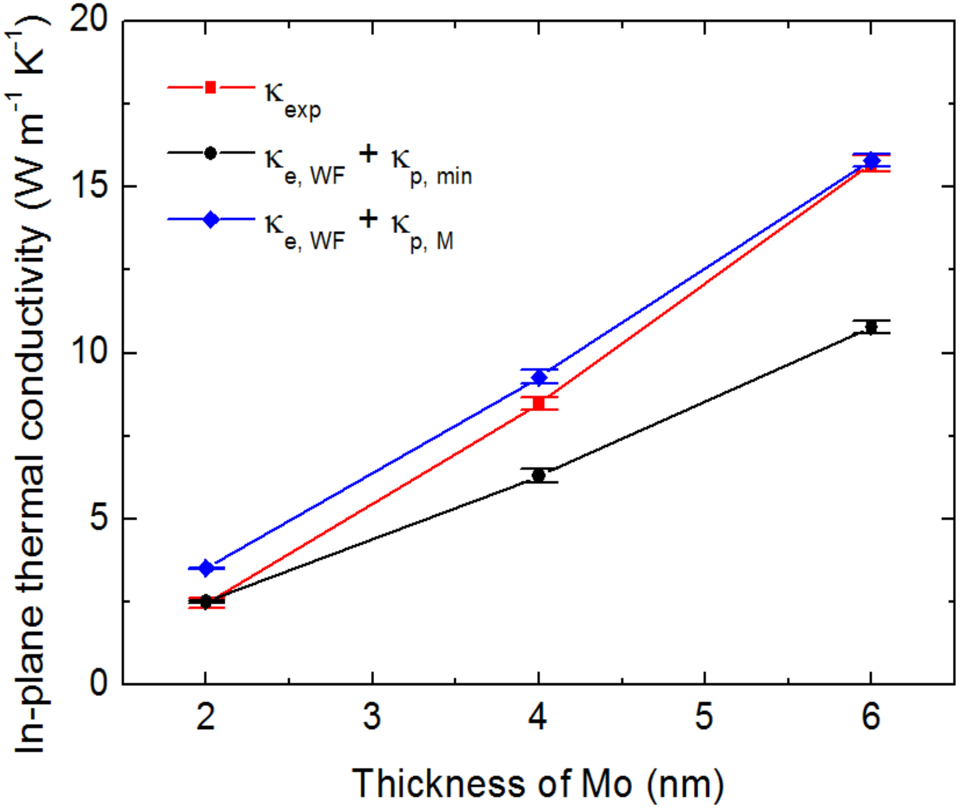
2. Matthiessen's rule

For nanostructured (polycrystelline) materials

$$\frac{1}{\lambda} = \frac{1}{\lambda_{Lattice}} + \frac{1}{\lambda_{Defects}}$$

# In-plane thermal conductivity

Measured data vs estimated electron + phonon contributions




- For 2 nm Mo minimum thermal cond limit gives better matching
  - For 4 and 6 nm Mo Matthiessen's rule rules